MELTING OF POLYMER BLENDS AND CONCOMITANT MORPHOLOGY DEVELOPMENT IN SINGLE SCREW EXTRUDERS

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Abstract - The current understanding of the melting stage in single screw extruders results from pioneering research efforts that were initiated in the fifties and continued for more than thirty years. Most of these theoretical and experimental studies used homopolymers as model systems, whereas in industrial practice there has been a considerable evolution in terms of the complexity of the materials being extruded. This work reports an attempt to monitor the melting sequence and the morphology development of immiscible physical and chemically compatibilized PA6/PP blends. A hybrid melting mechanism, incorporating elements of the Tadmor and of the Dispersive melting mechanisms seems to develop; the early stages of morphology development seem to be similar to those observed in the Haake mixer and Twin-screw extruder.

Introduction

Phenomenological studies of melting in single screw extruders were initiated in the fifties by Maddock (1959). Tadmor *et al.* (1966) developed the first mathematical description of Maddock's experimental observations. In his model, Tadmor assumes the existence of an elastic solid bed, formed by the compactation of the solid pellets, that is separated from the inner hot barrel wall by a thin melt film. The relative velocity between solids and barrel pushes this film into a melt pool, which grows near to the screw pushing flight. Simultaneously, the melt pool pushes the solid bed against the screw trailing flight, exposing more solids for melting at the solids-melt film interface. This sequence of events induces the progressive decrease of the solid bed width.

A rather different melting mechanism was observed by Zhu and Chen (1991) when using specific screw geometries, where dispersion of the solid particles in molten polymer was achieved by forcing the solid bed to break-up, followed by melting of these particles. Rauwendaal (1993) developed the first model of this dispersive melting mechanism, while Huang and Peng (1993) and Potente and Pape (2001) suggested different mathematical approaches to describe the same phenomena.

The above studies were performed using homopolymers, while complex systems are routinely processed nowadays. In the case of polymer blends, it has been shown that the melting stage is crucial to morphology development and, consequentially, to the final blend properties (Machado *et al.* (2006)).

A morphology evolution mechanism for the initial polymer blending stages was proposed by Scott and Macosko (1991) for batch mixers and confirmed by Sundararaj *et al.* (1992) for twin-screw extruders. However, and despite of some efforts (for example, Gosh and Tyagi (2002)), the morphology development in the case of single screw extruders is not clear.

This work reports a study of melting and morphology development of immiscible and compatibilized PA6/PP blends in a single screw extruder. The ultimate goal of the work is to correlate operating conditions and screw geometry with the morphologies that induce optimal properties.

Experimental

Materials

Polyamide 6 / Polypropylene (physical blends, PA6/PP and compatibilized *in situ* with Maleic anhydride grafted Polypropylene, PA6/PP/PP-g-MA), with different compositions, were selected as model systems. The specific grades (see Table 1) were chosen so that the viscosity ratio for the physical blends was approximately one. The melting temperature (from DSC thermograms) of PP and PA6 is 166ºC and 226ºC, respectively.

Table 1- Raw materials

Material	Trade name	Supplier
PP	ISPLEN 030 G1E	REPSOL
PP-g-MA	OREVAC CA 100	ARKEMA
PA6	Akulon F 130	DSM

Equipment

The blends were prepared in a prototype modular single screw extruder, with a diameter of 30 mm, L/D = 30 and a compression ratio of 2.5, fitted with

material sampling devices and pressure transducers along the barrel. A hydraulic contraption is available to extract quickly the screw in order to perform Maddock-type experiments. The screw profile can be modified using the same modularity approach as that of co-rotating twin screw extruders.

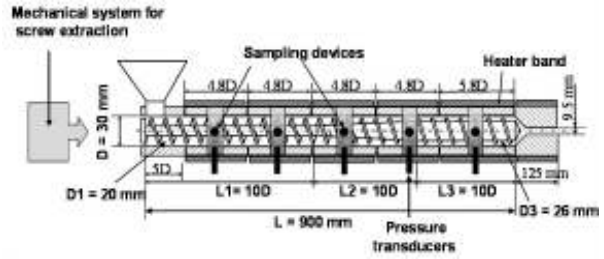

Figure 1- Extruder layout

Procedure

The blend components were pre-mixed in a drum mixer for 10 minutes before extrusion. After reaching steady state, the screw rotation was interrupted, the die was quickly removed and the screw was extracted from the barrel. This operation took approximately 1 minute. The polymer helix was removed from the screw and further cooled with compressed air. Cross-sections were cut from the polymer helices at regular down-channel intervals, immersed in epoxy resin for 24h, polished and immersed for one hour in a dyeing bath in order to tint PA6, thus facilitating its identification.
The melting sequence was studied using a stereoscopic magnifying glass, while the morphology evolution was observed by SEM (after freezing in liquid nitrogen and fracture). Samples were also submitted to selective chemical dissolution by formic acid or hot p-xylene to extract PA6 and PP, respectively.

Results and Discussion

A comprehensive study of melting of polymers blends must encompass various observation scales. In terms of melt development, Figure 2 shows that it is more complex than in the case of homopolymers.

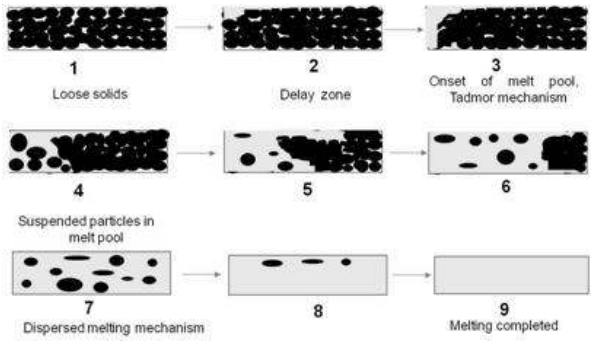

Figure 2- Hybrid melting mechanism of polymer blends.

If the early melting stages seem analogous to the conventional observations for homopolymers, from stage 4 onwards both the Tadmor and the dispersive melting sequences seem to develop concurrently. This evolution is valid for both the physical and compatibilized blends, although the duration of the intermediate steps depends on composition and operating conditions.
Monitoring the contribution of each blend component to the overall melting performance requires not only its detection, but also the quantification of its melting evolution. Figure 3 illustrates the role of each polymer in the melting sequence, taking into consideration the blend composition.

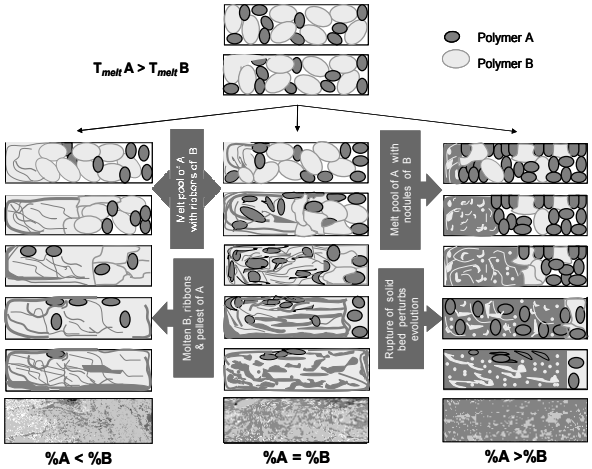

Figure 3- Melting mechanism of polymer blends

Not only the onset, but also rate of melting of each polymer is different. Therefore, it makes sense to investigate the corresponding morphological development. Figure 4 presents the type of data generated for this purpose. In this case, PA6 was extracted.

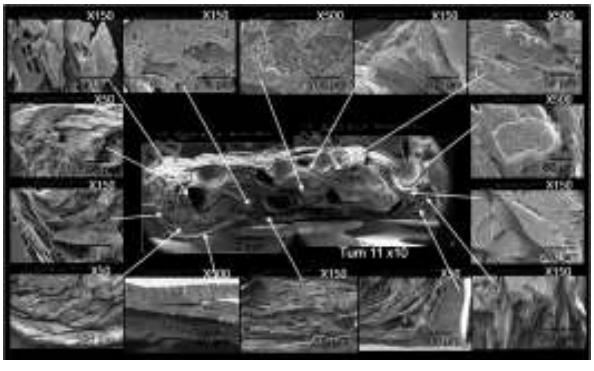

Figure 4- SEM micrographs at screw turn 11 for the PA6/PP 50/50 w/w blend.

The various micrographs across the channel depict different morphologies. Since the cross-channel flow

pattern in single screw extruder channels is well known, it is possible to recreate the global morphology evolution. Anyway, the individual photos of Figure 4 show structures that were reported by Scott and Macosko (1991).

Conclusions

The melting mechanism of polymer blends is more complex than that reported for homopolymers. A hybrid melting sequence, incorporating elements of the Tadmor and Dispersive melting mechanisms, seems to develop. The duration of the sequential steps is affected by blend composition and operating conditions. The blend components melt at different rates that depend again on blend composition, operating conditions, and location of the pellets in the channel. Morphological structures similar those reported for polymer blends processed in other types of equipment were observed upon melting. A rapid morphology evolution is preferentially observed at the solid pellets/molten material interface.

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